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Strain induced phase transformation in zirconium thin films

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ABSTRACT

While high temperature is known to transform zirconium from the hexagonal closed pack (hcp) to the body centered cubic (bcc) phase, there is little or no evidence of mechanical strain induced transformation to the face centered cubic (fcc) phase in the literature. We performed molecular dynamics simulation to show irreversible hcp to fcc phase transformation in zirconium, triggered at about 14.6% tensile strain. The transformation mechanism depends on the crystallographic direction of loading. Gliding of Shockley partial dislocations on prism plane $\{1\,0\,\bar{1}\,0\}$ is suggested facilitate this transformation. Nudged elastic band (NEB) theory was used to estimate the transformation energy barrier, which was observed to decrease with any increase in temperature.

1. Introduction

Zirconium is a transition metal with the hexagonal closed pack (hcp) lattice structure, which exhibits low thermal neutron absorption and good corrosion resistance at elevated temperature. Zirconium based alloys are commonly used as cladding material in nuclear reactors. In addition, its high melting point and biocompatibility makes zirconium an attractive material in aviation and surgical implant applications. High temperature (1135 K) is known to transform the phase from hcp to body centered cubic (bcc) [1]. Transition metals such as zirconium or titanium can exist in three phases such as hcp, bcc and face centered (fcc) [2,3], but hcp is mostly observed in zirconium [4,5]. Ji et al. [6] studied the metastable tetragonal states of zirconium using theory and experimentation. They conducted experiment to stabilize fcc zirconium using pseudomorphic epitaxy of zirconium films on the [0 0 1] surface of tungsten. The feasibility of fcc phase zirconium thin film was reported by Chopra et al. [7] and Hill et al. [8]. Chopra et al. [7] sputtered zirconium film on different substrates under vacuum condition. According to this study, room temperature deposition produced hcp zirconium, whereas deposition at 200–300 °C yielded fcc zirconium films up to 2-μm thickness. However, thicker films contain both the hcp and fcc phases. Hill et al. [8] studied the growth of zirconium films evaporated on a tungsten [0 0 1] substrate in ultrahigh vacuum condition and reported stable fcc structure up to 1800 K.

In this study, we investigate the effect tensile strain on phase transformation in zirconium thin films using Molecular Dynamics MD simulation. Molecular Dynamics (MD) simulation is an expedient computational tool for exploration of mechanical, thermal, chemical kinetics and phase transformation at the nano-scale. For example, phase transformation in metallic nanowires has been widely studied by this technique [9–11]. Existence of the fcc phase in zirconium has been suggested by atomistic simulation studies [12,13]. Computational investigation showed that the fcc structure could be formed near the crack tips and/or at the twin boundaries of the hcp structure [14,15]. However, strain induced hcp to fcc phase transformation in zirconium thin film is yet to be reported in the literature. We apply tensile load along two different directions to investigate the direction dependence of mechanical properties of hcp zirconium. In addition, we conducted our study at different temperatures to investigate the temperature effect on phase transformation. We also calculate the energy barrier for hcp to fcc phase transformation in zirconium using nudged elastic band (NEB) theory.

2. Computation details

We investigate phase transformation in zirconium thin films using the Embedded Atom Method (EAM) [16] potential. The functional form of the EAM potential is as follows:

$$E_{total} = \sum_{i=1}^{N} F_i(\bar{\rho}_i) + \frac{1}{2}\sum_{i=1}^{N}\sum_{i\neq j}^{N} \varnothing_{ij}(r_{ij}) \quad (1)$$

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$$\bar{\rho}_i = \sum_{j=1, j\neq i}^{N} \rho_i(r_{ij}) \quad (2)$$

where E_{total}, F_i, $\bar{\rho}_i$, $\varnothing_{ij}$ are the system total energy, embedded energy as a function of electron density, host electron density and pair potential as a function of distance r_{ij} between two atoms i and j respectively. In our simulation, a times step of 0.5 fs and 1 bar pressure was maintained. The simulation cell of hcp zirconium was built with dimension of 110 Å × 110 Å × 21 Å as shown in Fig. 1. Periodic boundary conditions were maintained in all directions. We minimized the energy using the conjugate-gradient (CG) method followed by the NPT dynamics for 0.1 ns using the LAMMPS [17] package. Fig. 1 shows the two loading directions in the specimen. Tensile loading was performed at the rates of 10^{10} s^{-1}, 10^{9} s^{-1}, 5×10^{8} s^{-1} and 10^{8} s^{-1}. Time integration during the loading was implemented using the NVT dynamics. Effect of temperature on phase transformation was studied at temperatures 10 K, 300 K, 500 K and 700 K. The mechanisms behind the observed phase transformation was studied using common neighbor analysis (CNA) and nudged elastic band (NEB) [18–21] simulation using LAMMPS. To perform NEB simulation, we require an initial and a final structure. We used hcp and fcc zirconium as our initial and final structure respectively. The model was validated by comparing elastic properties with the literature. Our calculated Young's modulus for zirconium thin film is 111 GPa along the [1 1 $\bar{2}$ 0] [11] loading direction which is in well agreement with existing literature value 109 GPa [22].

3. Results and discussion

Table 1 shows the lattice parameter (a and c) and cohesive energy (E_c) of the hcp zirconium thin film specimen calculated in this study using the EAM potential, which are in agreement with experimental and computational values in the literature.

After specimen preparation, tensile load was applied along the [1 0 $\bar{1}$ 0] and [1 1 $\bar{2}$ 0] directions, which correspond to x and y directions respectively as shown in Fig. 1. Fig. 2a shows the stress-strain behavior of hcp-zirconium under tensile loading along the x axis. The hcp phase remains stable up to about 15% strain, after which we observe a sudden drop in tensile stress accompanied by fcc phase transformation. Fig. 2b shows the mean squared displacement (MSD) during the tensile loading. Above 0.6 ns (corresponding to 14.6% strain) we notice a sudden peak in MSD curve as well as an increase in the slope. The sudden increase of MSD is due to the large deformation activated by the higher atomic mobility associated with the hcp to fcc phase transformation in zirconium.

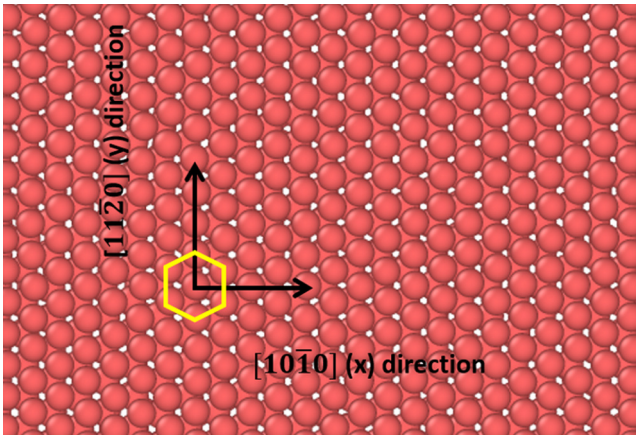

Fig. 1. Molecular dynamics simulation cell of hcp zirconium (top view from [0 0 0 1] plane).

Table 1
Comparison of zirconium properties.

Properties	Experiment [23,24]	DFT [25]	Present study EAM [16]
a (Å)	3.23	3.23	3.227
c (Å)	5.15	5.17	5.147
E_c (eV/atom)	−6.32	–	−6.33

Fig. 3 shows the atomic stress distribution on zirconium thin film during the tensile loading. Initially, the zirconium atoms were nearly stress free in the hcp phase as shown in Fig. 3a. However, atomic stress increases with the increment of tensile strain, and the hcp zirconium transforms to fcc phase around 14.6% strain (Fig. 3b). Failure initiates at about 24.9% strain through nucleation of cracks that relieve the local atomic stress as shown in Fig. 3c.

Phase Transformation Mechanism: Fig. 4 shows the mechanistic roadmap of the hcp to fcc phase transformation in zirconium. Some of the atoms are shown in a different color to highlight the process. Fig. 4a shows the crystal structure at 14.6% strain, which is the onset of the phase transformation. Here, the phase is still hcp and the {1 0 $\bar{1}$ 0} prism plane as a dislocation glide plane, which could be the primary glide plane for hcp metals with less than 1.63 c/a ratio [26]. After the hcp to fcc phase transformation, we notice that it follows the Burger notation i.e. [1 1 $\bar{2}$ 0]|| [1 1 0] and [0 0 0 1]||[0 0 1], which is similar to the experimental study [27]. We analyzed the simulation trajectory using the DXA algorithm [28] to identify the dislocations during the phase transformation. We noticed that the phase transformation is primarily attributed to the Shockley partial dislocations with a Burgers vector $\bar{b} = a/6[1\,1\,\bar{2}\,0]$. Recent experimental [27] and computational [29] studies also report similar type of phase transformation mechanism in hcp titanium. Fig. 4b shows the intermediate stage. Common neighbor analysis (CNA) shows presence of both bcc and fcc structures due to the strain induced twinning and stacking faults. Fig. 4c shows the lattice structure almost (98.8%) transformed the fcc phase.

Fig. 5a shows the energy plot during the NEB simulation. We used 20 replica including initial hcp and final fcc zirconium structure. The lattice constants were, a = 3.227 Å with a c/a = 1.594 for hcp and a = b=c = 4.537 Å for fcc phases respectively. We performed multiple NEB simulation run using different spring constant starting from 5.0 eV/Å. Fig. 5b–g show the phase transformation including intermediate structure during the NEB simulation. Reaction coordinates 0.0 and 1.0 correspond to the hcp and fcc structures respectively. The energy barrier plot indicates that hcp-fcc phase transformation requires additional stimuli, which is shown to be provided via tensile straining. It is important to note that 14.6% strain is very high for experimental conditions since real materials contain defects that lead to plastic deformation before the phase transformation can take place.

Failure Mechanism: Starting with the pristine crystal, we did not observe any significant dislocation nucleation activity before phase transformation. Fig. 6a shows a complete hcp to fcc phase transformation above 14.6% strain. The specimen fails at 24.9% strain as shown by Fig. 6c. Prior to failure, the structure undergoes hcp, bcc and fcc structures. In this present study, we use the DXA algorithm [28] to investigate dislocation dynamics during tensile loading. Fig. 6d show dislocation nucleation due to the plastic deformation after the phase transformation. The observed dislocations were both edge (deep green color arrow) and screw (red color arrow) types. The Burger vectors are shown by cyan color arrow in Fig. 6a and b. Fig. 6b shows {1 1 $\bar{2}$ 0} twinning (along edge dislocation line extending from left to right side of simulation cell) prior to failure due to the plastic deformation of zirconium. Fig. 6c shows